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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: ]  
Mats SUNDBERG et al. ] Group Art Unit 3742  
Serial No.: 10/510,145 ]  
Filed: April 19, 2005 ]  
For: METHOD OF MAKING A HEATING ] Examiner: S. Ralis  
ELEMENT OF THE MOLYBDENUM [  
SILICIDE TYPE AND A HEATING ]  
ELEMENT ]

**DECLARATION UNDER 37 CFR 1.132**

Commissioner for Patents  
P. O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Mats Sundberg, hereby declare as follows:

1. That I am one of the inventors named in the above-identified application;  
2. That in regard to the issue of heating elements spalling or not influenced by the purity of the raw materials from which the heating elements are formed, tests were performed under my supervision at Kanthal AB, Hallstahammar, Sweden, to assess the adhesion and integrity of surface layers of  $\text{Al}_2\text{O}_3$  on molybdenum silicide heating elements under thermal cycling conditions. The tests involved  $\text{Mo}(\text{Si}_{1-x}\text{Al}_x)_2$  heating elements having different amounts of  $\text{SiO}_2$ . Thermal cycling tests were run using one group of  $\text{Mo}(\text{Si}_{1-x}\text{Al}_x)_2$  heating elements that included bentonite, a material containing about 60%  $\text{SiO}_2$  by weight, and another group of  $\text{Mo}(\text{Si}_{1-x}\text{Al}_x)_2$  heating elements in which substantially 98% pure  $\text{SiO}_2$  was substituted for the bentonite;

3. In a first example, 4 pcs of 3mm/6mm thickness heating elements in which Mo(Si<sub>1-x</sub>Al<sub>x</sub>) was utilized as a component of the heating element composition and wherein the value of x was about 0.9. A bentonite composition was used as an additive to form the heating element and contained the following constituents and amounts by weight (60% SiO<sub>2</sub>, 23% Al<sub>2</sub>O<sub>3</sub> + Mg, Ca Na, and Fe-oxides to total 100%). The heating elements were run in a high-alumina, insulated laboratory furnace for 30 heating cycles between room temperature and T=1575 °C with a 4 hr dwell per heating cycle (2 hr heating rate and natural cooling). The thermal cycling resulted in spalling and flaking of the outer layer of Al<sub>2</sub>O<sub>3</sub>, and it also resulted in the formation of pits on the surface of the heating element.

4. In a second example, heating elements were prepared in which Mo(Si<sub>1-x</sub>Al<sub>x</sub>) was again utilized as a component of the heating element composition and wherein the value of x was about 0.9. Instead of bentonite having an SiO<sub>2</sub> content of about 60% by weight, as in the first example of paragraph 3 above, SiO<sub>2</sub> powder that was specified as containing a minimum SiO<sub>2</sub> content of 98% was used as the additive in the place of the bentonite that was used in the first example. The heating elements were subjected to the same thermal cycling running conditions as in the first example of paragraph 3 above, but for 92 heating cycles. The resulting Al<sub>2</sub>O<sub>3</sub> oxide scale that was formed remained adhered to the heating element surfaces and was both smooth and dense, and it did not flake off the surfaces of the heating elements.

5. In a third example, a heating element was prepared having the composition of the second example in which the Mo(Si<sub>1-x</sub>Al<sub>x</sub>) component had the same value of x of 0.9, and the element included SiO<sub>2</sub> powder having a purity of at least 98%. The

heating element was energized in open air to attain a temperature of 1580°C that was maintained for 1100 hours. A thin and adherent oxide outer layer was formed.

6. In a fourth example 6 pcs of heating elements in which the Mo(Si<sub>1-x</sub>Al<sub>x</sub>) component had the same value of x of 0.9, and included SiO<sub>2</sub> having a purity of at least 98% as in the second and third examples above were formed with a 6 mm heating zone. The heating elements were installed in an atmospheric furnace (CM Furnaces Inc.) and were run under an argon flow of 4.5 liters/min with a heating element temperature of 1560°C (the furnace temperature was 1500°C). The heating elements were inspected after cooling down every 7 days. The oxygen content (30-50 ppm) and dew point (-40 °C) were low, but were sufficient to form Al<sub>2</sub>O<sub>3</sub> on the surfaces of the heating elements from residual oxygen. After 41 days operation, the heating elements came out in excellent condition with an adherent, dense oxide outer layer and without peeling.

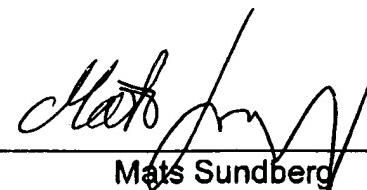
7. In a fifth example, a 4-shank 6 mm heating element in which the Mo(Si<sub>1-x</sub>Al<sub>x</sub>) component had the same value of x of 0.9, and included bentonite having the composition of the bentonite of the first example of paragraph 3 above, was formed with a 6 mm heating zone. The heating element was heated in air at a temperature of 1400°C for one week (7 days x 24 hours/day). The heating element rested on a high-purity, alumina refractory brick inside the furnace structure whose hot face insulation consisted of all alumina. After cooling down to room temperature severe outer surface layer Al<sub>2</sub>O<sub>3</sub> spalling occurred. The oxide scale was porous and extremely fragile. Chemical analysis of the oxide scale showed high levels of CaO and Na<sub>2</sub>O that originated from constituents of the bentonite additive, and which had diffused from the

material within the body of the heating element to the outer scale layer.

8. That the undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and, further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of any patent that issues from the present application.

18.02.09

Date



Mats Sundberg